# RADIATION PHYSICS NOTE 109

## Fermi National Accelerator Laboratory
## Radioactivity Release Criteria for Materials, Equipment and Waste

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## I. Introduction

It is the intent of this document to provide the criteria and rationale which will form the foundation for decisions by Fermilab personnel regarding the proper radioactive classification of materials, equipment and waste released from the Fermilab site. A summary of Fermilab's policies and procedures for determining whether a material or equipment item is radioactive is given herein. Specific procedures are maintained as separate documents referenced as appropriate.

Except for the case of bulk or volume contaminated solids, DOE Order 5400.5 specifies adequate criteria for the release of materials and equipment from DOE facilities. This order explicitly considers both fixed and removable surface contamination. Guidelines pertaining to volume or bulk contamination of aqueous solutions and air are also set forth in the Derived Concentration Guides of Chapter III. For several years, this order was used as the basis of Fermilab's release criteria for measurements performed on aqueous solutions with analytical laboratory instrumentation. Policies coming out of a DOE imposed moratorium in May of 1990 on hazardous waste shipments have made it necessary to abandon such "de minimus" based release criteria.

Materials and equipment are considered to be *potentially* contaminated or activated if they enter or are removed from a Radioactive Material Management Area (RMMA). These areas are defined in the Fermilab Procedures for Establishment and Management of Radioactive Material Management Areas.[1] Materials and equipment removed from other areas at the Laboratory are not considered to be potentially contaminated or activated. This release criterion applies to all solids and contained liquids which are removed from a RMMA or which have unknown origin.

## II. Definitions

**Volume contamination:** Volume contamination refers to material that has been contaminated in depth with radioactivity through the process of activation, such as, for example, materials activated by particle beams. These materials can take the form of solids or contained liquids.

**Bulk contamination:** Material that has been contaminated in depth with radioactivity through the process of mixing radioactivity into virgin material. For example, runoff from a cleaning fluid used to clean an activated magnet may be bulk contaminated through removal of surface contamination from the magnet. These materials can also take the form of solids or contained liquids.

**Radioactive material:** For the purpose of material (including waste) and equipment management at Fermilab, an item is considered radioactive if:

1. It contains removable surface radioactivity which equals or exceeds the limits established in DOE Order 5400.5.

   or

2. It contains volume or bulk accelerator produced radioactivity which produces a count rate on either a Bicron Analyst™ or a Victoreen Thyac™ and on an Eberline E140N or Ludlum 177-4 Frisker survey instrument that is statistically distinguishable from the measurement detection limits at a 95% confidence level. An equivalent way to state this criteria is that the item produces a count rate on either a Bicron Analyst™ or an Eberline E140N Frisker survey instrument that is statistically distinguishable from background at a 99.998% confidence level.

   or

3. It contains radioactive isotope concentrations which exceed the limits established in DOE Orders or guidance.

## III. Quantification of the Release Criteria for Survey Instruments

### A. Surface Contamination

Immediately upon removal from a RMMA, all items are contact surveyed with a Ludlum 177-4 or an Eberline E140N Frisker survey instrument. These surveys must be conducted in accordance with established Fermilab procedures as set forth in reference 2. The Ludlum 177-4 and the Eberline E140N Frisker have a 5.08 cm. diameter Geiger-Mueller counter with a thin window (1.5 mg/cm$^2$) for their detector. They have excellent $\beta$ sensitivity but are less than 1% efficient in detecting $\gamma$ rays. If any area on the item exhibits a count rate statistically different from the mean background count rate on the Eberline E140N or the Ludlum 177-4 survey instrument at a 99.998% confidence level, then that item must be considered as suspect surface contaminated material. This criteria is conservatively implemented by considering any item exhibiting 50 cpm or greater above ambient background as suspect radioactive material. If decontamination of this item is unsuccessful in reducing the count rate below 50 cpm above background then the area of highest count rate must be wiped for surface contamination according to established Fermilab procedures.[2] Items are only released for unrestricted use when the removable radioactivity levels are below the pertinent Surface Contamination Guidelines given in Figure IV-1 of DOE Order 5400.5.

Surface contamination is quantified by taking a wipe survey of the suspected material. These surveys must be performed in accordance with established Fermilab procedures.[2] The wipes are subsequently counted on a gas-flow proportional counter system operated within the ES&H Section by personnel trained in its proper use and in accordance with written procedures.[3] In accordance with DOE Order 5400.5, the release criterion is 1000 dpm/100 cm$^2$ wipe or about 0.5 nCi/100 cm$^2$ wipe for normal $\beta/\gamma$ emitters. There are no $\alpha$–emitters in the normal Fermilab activated material and waste streams, so that the $\alpha$–emitter categories mentioned in 5400.5 are not relevant.

## B. Volume or Bulk Contamination (Residual Radioactivity)

The order defines "volume contamination of material" as material contaminated in depth such as "...activated material or smelted contaminated metals." No smelted materials are used at Fermilab but cleaning operations can result in bulk contamination of the cleaning residue. The only items at Fermilab subject to "volume or bulk contamination" are materials and equipment which become radioactive when exposed to the beam or beam spray or residue from cleaning operations on activated material, radioactive sources, or the uranium plates which are used in the D0 calorimeter, and occasionally elsewhere. Included among the areas in which activation may occur are areas posted as a radiation area or higher (e.g., high radiation area or very high radiation area) hazard area in accord with reference 1. Areas where bulk contamination can occur are defined by the division/section Radiation Safety Officers (RSOs) in accordance with the directives found in reference 1. For solid objects, in the absence of removable contamination regulated by the above surface contamination release criterion, external radiation exposure from gamma-emitters is the most significant exposure pathway. Although the possibility of subsequent incineration of released material is highly improbable, it has been considered below for the sake of completeness.

DOE Order 5400.5 does not set forth specific criteria for volume or bulk contaminated materials. Hence it is necessary for Fermilab to establish a practical release criterion for materials, equipment, and waste based on the use of field survey instruments. These criteria are a conservative implementation of the definition set forth in Section II of this document. Materials, equipment, or waste items are considered to be radioactive if:

- On a contact survey the gross count rate with any surface is equal to or greater than twice the mean background count rate as measured in a low background (<2000 cpm) with a standard Bicron Analyst™ or Victoreen Thyac™ gamma scintillation probe on the X10 scale.

or

- On a contact survey the net count rate above mean background with any surface is greater than 2000 cpm as measured in a moderate background (≥2000 cpm but <3000 cpm) with a standard Bicron Analyst™ or Victoreen Thyac™ gamma scintillation probe on the X10 scale.

or

- On a contact survey the net count rate above mean background with any surface is greater than 50 cpm above mean background on an Eberline E140N or Ludlum 177-4 Frisker survey instrument.

These criteria are well below the instrument detection limits at a 95% confidence level and a measurement statistically different from background at a 99.998% confidence level.[4] Persons who make these determinations receive special, documented training in survey techniques and proper use of the instrument. Items found to be radioactive under this definition are labeled appropriately and not released for off site shipment unless they are specifically treated as shipments of radioactive materials to authorized recipients (i.e., other DOE facilities, or organizations possessing appropriate USNRC agreement state licenses). The Bicron Analyst™ survey instrument uses a 1.5 inch (3.81 cm) diameter by 1 inch (2.5 cm) long NaI(Tl) scintillation counter and the Victoreen Thyac™ survey instrument uses a 1.0 inch (2.5 cm) diameter by 1.5 inch (3.81 cm) long NaI (T1) scintillation counter. Such

devices are reliable survey meters for this purpose because of their high sensitivity to gamma radiation. At Fermilab, natural background typically induces a count rate varying from 1000 to 3000 cpm on the Bicron Analyst™ and Victoreen Thyac™ survey instruments. At a specific location, background typically varies by only ±200 cpm. Therefore when combined with a preliminary battery check as specified in training provided to instrument users, this provides a convenient check that the instrument is functioning properly.

## IV. Potential Consequences of this Release Criteria

Studies conducted with radioactivated materials and calibrated γ-emitting radioactive sources with energies between 0.5 and 1.3 MeV have indicated that 2000 counts above background on a Bicron Analyst™ or Victoreen Thyac™ instrument corresponds to exposure rates between 0.006 and 0.012 mR/h with an average of about 0.01 mR/h (Ref. 4). Reasonably long-lived radioactive isotopes produced at high-energy accelerators (such as those at Fermilab) that constitute the nuclides in the equipment and material released are mostly $^{3}H$, $^{7}Be$, $^{22}Na$, $^{54}Mn$, and $^{60}Co$. Except for $^{3}H$ these isotopes are gamma ray emitters with energies between 0.478 keV and 1.33 MeV. (See Ref. 5 for further discussion of $^{3}H$ in solid materials.) The Bicron Analyst™ and Victoreen Thyac™ survey meters mentioned above are particularly sensitive to such energies of emission. Ref. 5 documents that 2000 net counts, or 0.010 mR/hr, successfully detects point sources and screens drums of material containing accelerator produced isotopes at levels well below the USNRC exempt limits and concentration values. While these limits and concentrations are not necessarily germane to current DOE guidance, they do provide useful benchmarks for purposes of comparison in view of the lack of specific criteria in DOE 5400.5 for volume or bulk contaminated materials.

At the detection criteria of 2000 cpm net count rate, if all the counts were due to the presence of added radioactive materials rather than a statistical variation in background, this could result in a minimum detectable specific activity of between 20 and 50 pCi/g (depending on the gamma ray energy) in bulk materials such as aluminum, concrete, or iron if irradiated uniformly (Ref. 5). These concentrations are far less than the USNRC exempt concentrations (10 CFR § 30 Schedule B) for listed accelerator produced radionuclides such as $^{54}Mn$, $^{60}Co$ and $^{22}Na$. Even for $^{7}Be$, an accelerator produced radionuclide with only a 10% gamma ray emitting branch, the adopted criterion is sensitive enough to assure detection at a concentration above 500 pCi/g (Ref. 5), well below the guidelines.

If the bulk material is not irradiated uniformly (e.g., a magnet in an enclosure struck by a pencil-like particle beam from the Tevatron), the entrance activation level is within a factor of two of the maximum value, which occurs at a depth of 1.5-3.0 feet within the material, and at a radial distance of 0-3 cm from the point of irradiation. This is documented in Figs. VIII.8 and VIII.25 of Ref. 6 (reproduced in Figs. 2 and 3 of Ref. 5) which, for 1000 GeV protons, show the number of stars/cm$^3$ (a quantity directly related to the amount of residual radioactivity) as a function of depth in iron and concrete. Thus, even under such conditions, a 2000 cpm net count rate is sufficient to detect residual radioactivity levels less than the exempt concentrations. (The nonuniformities of this type are maximized at the highest available proton energy at Fermilab of 1000 GeV.)

The possibility that bulk material released under the less than twice background criterion of 2000 cpm net count rate could subsequently be subjected to incineration has been investigated in Ref. 5. Fig. 5 in that reference shows that at most 22% of the Derived

Concentration Guide (DCG), given in DOE Order 5400.5, for the most restrictive accelerator produced radioisotope $^{60}Co$, will be released at ground level from a plume downwind of the furnace stack. This number is, furthermore, an overestimate since DCG's are based on a committed dose equivalent of 100 mrem taken into the body during a full year for an individual exposed to such a concentration during the whole time, while any reasonable estimate of the burning of bulk material should probably be measured in terms of hours or days. For example, a massive magnet (weighing 35,000 pounds) might take seven hours to burn (Ref. 5). An individual at the worst downwind location would experience about 0.02 mrem during this time.

It is observed that DOE Order 5400.5 itself requires that average levels of $\gamma$-radiation, within any building to be released without restrictions, be less than 0.020 mR/h. The most liberal element of the present criterion, <.010 mR/hr, is a factor of 2 lower than this value, and is therefore sufficient to assure compliance with this requirement. It is noted furthermore that this release criterion corresponds to an absorbed dose rate associated with surface contamination that is a factor of 20 less than the 0.2 mrad/h at 1 cm due to $\beta/\gamma$ emitters specified in Table IV-1 of the order.

Although the criterion under discussion cannot detect $^{3}H$ in bulk matter nor in contained liquids, its presence in matter is comparable to that for other isotopes to which the screening criterion is sensitive (Ref. 5). In solids, $^{3}H$ will be bound in the material, and thus does not present any hazard potential. For contained liquids (e.g., hydrocarbons, water) irradiated for periods up to a year by primary and secondary high-energy beams, the activity associated with $^{7}Be$, to which the criterion is sensitive, will be larger than that for $^{3}H$ even after 8-9 months of post-irradiation "cooling." Thus, for most such contained liquid samples the criterion is appropriate to screen the material for radioactivity. After very long cooling times however sampling and analysis for $^{3}H$ may be the only method for assessing levels of such activity in contained liquids. It should be noted that the USNRC exempt quantity and exempt concentration limits, as well as the DCG limit from DOE Order 5400.5, are significantly larger than similar guidelines for other radioisotopes so that any $^{3}H$ component does not represent a significant exposure pathway (Ref. 5).

## V. Release Criteria and Analytical Instrumentation

On rare occasions, it will become necessary to screen materials or representative samples of materials for volume/bulk radioactivity using analytical instrumentation. Generally both gamma ray spectroscopy and liquid scintillation counting (LSC) analysis must be performed on such samples. In both cases the analysis will be conducted according to procedures set forth in reference 7 using a 1 hour count time for gamma ray analysis and a 1.5 hour count time for LSC analysis. In all measurements, the material will not be considered radioactive unless radionuclides are detected in the sample at levels exceeding the limit of detection for the particular instrument used to make the measurement at a 95% confidence level. This is the only way to insure that the spectroscopist is not placed in a situation where he must quantify radionuclides at confidence levels between 50 and 95%.

## VI. Survey Techniques and Procedures

Persons who perform surveys for off site release of materials receive performance based training in Fermilab procedures and techniques for waste handling and screening. This training is provided by the ES&H Section and is documented in the Laboratory training data base. A list of persons authorized to perform such surveys is updated at frequent

intervals. Procedures for the performance of surveys for both surface contamination and volume or bulk (residual) radioactivity are documented in reference 2.

## A. Surface Contamination

As mentioned, surface contamination is determined by taking a wipe survey of the suspected material. A standard wipe is performed by using a piece of cloth or paper and wiping an area of 100 $cm^2$ (4" by 4"). Details of this process can be found in reference 2. It is then counted on a Tennelec LB5100 Series 2 counting system that consists of a 100 sample capacity automated changer mechanism and two detector assemblies—a 2.25" (5.72 cm) thin window (80 $\mu g/cm^2$) gas flow proportional counter and a 2" (5.08 cm) by 2" by 2" NaI detector. The assembly is operated within the ES&H Section by personnel trained in its proper use under documented procedures for use, maintenance and calibration (Ref. 3).

The efficiency for detection of standard $\beta/\gamma$ emitters typical of those radionuclides produced at an accelerator varies from 54 cpm/nCi for $^{54}Mn$ to 567 cpm/nCi for $^{22}Na$ (Ref. 8). The efficiencies are determined for each isotope deposited on a cloth wipe and attached to a typical Fermilab aluminum planchet. An average wipe of accelerator produced isotopes is detected with a sensitivity of about 400 cpm/nCi. The background on the counting system used (see above) is 1-2 cpm when used for $\beta/\gamma$ analysis.

## B. Volume Contamination

The Bicron Analyst™ survey instrument uses a 1.5 inch (3.81 cm) diameter by 1 inch (2.5 cm) long NaI(T1) scintillation counter. Such a device is a reliable survey meter for this purpose because of its high relative sensitivity to gamma radiation above 200 keV in energy. A NaI(Tl) scintillation crystal of this size has an absolute efficiency for 0.5-1.0 MeV gamma rays that is 3.5 to 5 times that for a standard GeLi solid-state counter on contact (or at 1 cm distance) with a radioactive sample. The lower level discriminator on the Victoreen Thyac™ survey meter is set at the factory and the lower level discriminator on the Bicron Analyst™ is set to match the Thyac reading in the $\gamma$ field generated by a 0.1 $\mu Ci$ $^{137}Cs$ source.

Backgrounds at Fermilab are typically between 1000 and 3000 cpm as recorded with either the Victoreen Thyac™ or Bicron Analyst™ survey meters. With the Bicron Analyst operated on the X10 scale as required in the stated release criterion (see above), it responds in 1 second to 90% of its final reading. Based on a 1 sec response (90% full scale) time, an analysis[4] which incorporates some of the more salient random errors in the measurement demonstrates that the minimum net counting rate that can be detected at the 95% confidence level averages around 2000 cpm for typical background rates. In low background areas, for example in a 1000 cpm mean background, the Fermilab release criteria becomes 1000 net counts which is about 40% lower than the minimum detectable quantity of radioactive material that can be reliably (with 95% confidence) detected in that background. Hence the Fermilab release criteria is a conservative implementation of the radioactive definition found in Section II of this document.

Due to the occasional existence of the $^{195}Hg$ isotope and its daughters, $^{195}Au$ and $^{195}Pt$, on material stored near the Neutron Therapy Facility (NTF) target, a Geiger-Mueller pancake survey detector (Frisker), such as is included on the Ludlum 177-4 or the Eberline E140N Frisker portable instrument must also be used to screen material and equipment before it can qualify for unconditional release. This decay chain is not efficiently detected with the

Victoreen Thyac™ or Bicron Analyst™ survey instruments as a result of the low energies and branching ratios for the emitted gamma rays.

In rare cases when the material to be released has residual magnetism, only a Geiger-Mueller pancake survey meter can be used to screen the material. This phenomena is due to deflection of the electrons in the photomultiplier tube and is readily identified by either spurious "full scale" or "zero" readings even in a relatively weak magnetic field. The efficiency of the GM tubes in these survey instruments is constant at energies between about 0.15 and over 1.2 MeV. Recent studies (Ref. 9) have shown that a count rate of 2000 cpm net on a Bicron corresponds to 65-75 cpm net on the Frisker survey meters for accelerator-activated materials. Typical backgrounds vary from 30 to 50 cpm on Frisker survey instruments.

For backgrounds from 1000 cpm to 2500 cpm $L_d'$ (ref. 4) will range from 1700 cpm to 2300 cpm net count rate. These numbers are fully consistent with Fermilab's present release criteria, with current DOE moratorium guidance, and with realistic measurement detection limits.

The ES&H Section at Fermilab is responsible for maintenance, calibration, and repair of radiation monitors—both portable and area. Written calibration procedures for each instrument exist. For the Thyac, Bicron and Frisker type survey instruments a calibration is performed at least annually in accord with standards specified in DOE orders.

## References


1. Fermilab Radiological Control Manual, most recent revision, first issued 4/1/93.

2. Fermilab Waste Handling and Certification Procedures Manual - Current Revision.

3. F. P. Krueger, Operation Of The Tennelec LB5100 Sample Changes System, Radiation Physics Note No. 53, (Fermilab, Batavia, IL, Revised October 1990).

4. V. R. Cupps, Survey Instrument based Release Criteria and DOE Policy, Radiation Physics Note 108, 5/93.

5. J. D. Cossairt and A. J. Elwyn, Response of 1 1/2" by 1" NaI(Tl) With Respect To A Release Criterion, Radiation Physics Note No. 87, (Fermilab, Batavia, IL, Revised October 1991).

6. A. Van Ginneken and M. Awschalom, High Energy Particle Interactions in Large Targets, Vol. 1, (Fermilab, Batavia, IL, 1975).

7. Fermilab Activation Analysis Laboratory Procedures and Policy Manual, Revision 1, 6/93.

8. F. P. Krueger, Calibration of the Tennelec LB5100 and APC Wipe Counting Systems, Radiation Physics Note No. 96 (Fermilab, Batavia, IL, May 1992).

9. F. P. Krueger and A. J. Elwyn, Comparison Of The Response Of Bicron and Frisker Survey Instruments, Radiation Physics Note (to be completed).

10. Accelerator Division Safety Manual, (Fermilab, Batavia, IL, 1988).

11. J. Shapiro, Radiation Protection (Harvard University Press, Cambridge, MA, Third Edition, 1990), p. 255.

12. J. C. Lochamy, The Minimum Detectable Activity Concept, NBS SP456 (National Bureau of Standards 75$^{th}$ Anniversary Symposium, 1976), and PSD #17, EG&G ORTEC System Application Studies.

13. M. Barbier, Induced Radioactivity, John Wiley & Sons, Wiley Interscience Division, New York, New York, 1969.

14. V. McLane, C.L. Dunford, P.F. Rose, Neutron Cross Sections, Volume 2, Neutron Cross Section Curves, National Nuclear Data Center, Brookhaven National Laboratory, Academis Press, Inc., New York, New York, 1988.